EXPERIMENTAL EFFORTS AND RESULTS IN FINDING NEW HEAVY SCINTILLATORS*

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Abstract

New heavy scintillators are being discovered with increasing frequency. In recent years NaI(Tl) (with its high light output and energy resolution) has been joined by BGO (with its high stopping power), BaF_2 (with its excellent timing resolution), and CeF_3 (with its speed and short Molière radius). More than 10 potentially useful scintillators have been under development in the past five years, such as $PbSO_4$ and Lu_2SiO_5(Ce). We tabulate the characteristics of these and other scintillators, including wavelength, luminous efficiency, decay time, and initial intensity. We describe a search strategy and the prospects for finding the “ideal” heavy scintillator, which would combine the light output of NaI(Tl) and CsI(Tl), the stopping power of BGO, and the speed of BaF_2 and ZnO(Ga).

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1. Introduction

The widespread use of heavy scintillators was initiated in the late 1940's when Robert Hofstadter and co-workers developed NaI(Tl) and demonstrated the use of early photomultiplier tubes to detect scintillation flashes[1-4]. In the 1950's the same research group discovered scintillation in CsF[5] and in pure NaI at 77K[6]. Over the years a number of other useful scintillators have been discovered and developed, and summaries of their properties may be found in references 7-14.

For gamma ray detection and spectroscopy, heavy inorganic scintillators are most often used. In this role, scintillators have a number of desired characteristics:

- The stopping power (the probability of complete absorption of the photon energy), which is enhanced by choosing a high density and high atomic number
- The timing resolution, which is enhanced by a short decay time and a large light output
- The energy resolution, which is enhanced by a large light output
- The dead time, which is reduced by a short decay time
- The wavelength of emission, which should be matched with the spectral response of the photodetector
- Mechanical ruggedness
- Radiation hardness
- Chemical stability in normal atmospheric conditions
- Availability of large, clear crystals at low cost

As summarized in the following section, many scintillators are available, but none excels in all the above properties. As a result, choosing a scintillator requires judicious compromises. In subsequent sections we list both established scintillators, those under development, and candidate compounds that hopefully will be developed to widen the selection of choices.

2. Established Heavy Scintillators

Table 1 lists established heavy scintillators that are either currently or have been in commercial production. Heavy-atom scintillators for gamma-ray detection have been emphasized, but others have been included for reference. For gamma-ray detection, the following are of particular note

- High luminous efficiency (in photons / MeV): NaI(Tl) and CsI(Tl)
- High density, high atomic number, and short gamma-ray attenuation length: BGO
- Short Molière radius: BGO, CeF_3

- High initial photon intensity (in photons / MeV / ns) and an excellent timing resolution: BaF_2
- High luminous efficiency and wavelength suitable for silicon photodiodes: CsI(Tl), $CdWO_4$

3. Heavy Scintillators in Limited Availability

Table 2 lists scintillators that have been studied as single crystals, and whose properties as scintillation detectors have been measured, but are not yet in large-scale commercial production. In the case of $PbCO_3$ and $PbSO_4$, the largest clear crystals available are small samples of natural minerals.

4. Interesting Heavy Compounds not Available as Scintillation-Quality Crystals

For several years we have been using pulsed synchrotron x-radiation to measure the radioluminescence of over 400 compounds in powdered form[65, 66]. The technique is able to make accurate measurements of wavelength and decay times and approximate measurements of luminosity. The decay timing spectrum is measured using the delayed coincidence method of Bollinger and Thomas[67]. Using these methods, we have discovered (or rediscovered) x-ray excited fluorescence from $PbWO_4$, CeF_3, $PbCO_3$, $PbSO_4$, Yb_2O_3, CuI, $BaCl_2$, and $CeCl_3$ in powdered samples (Table 3). Subsequently, we were able to acquire synthetic crystals of $PbWO_4$ and CeF_3, and natural crystals of $PbCO_3$ (cerussite) and $PbSO_4$ (anglesite).

Recently, we have described the design of a table-top pulsed x-ray system for this work[73]. It uses a laser diode, a light-excited x-ray tube, and a microchannel phototube. The measured system timing resolution is 109 ps fwhm.

It is interesting to note that in 1947 Robert Hofstadter discovered the high luminosity of NaI(Tl) before growing a crystal. He produced a molten glaze of NaI and Tl halide and placed it on a photographic plate along with samples of anthracene, naphthalene, KI(Tl), NaCl(Tl), KBr(Tl), $CaWO_4$, etc. After exposure to a radium source, the developed film was blackened by the NaI(Tl) powder to a much greater intensity than any of the other samples[1, 2].

Table 1 Properties of Established Heavy Scintillators[a]

	density (gm /cm^3)	μ^{-1} b	hygro-scopic	λ_{max} (nm)	index refr.	photons /MeV	decay time (ns)	photons /MeV /ns	refs
anthracene[c]	1.25	8.79	no	450	1.62	16,000	30	550	11
BaF_2	4.89	2.29	no	195, 220	1.49	1,800	0.8[d]	3,000	13, 15-20
				310		10,000	630	15	
$Bi_4Ge_3O_{12}$	7.13	1.11	no	480	2.15	700	60	12	13, 21-25
				480	2.15	7,500	300	25	
					totals =	8,200	total =	37	
$Bi_4Ge_3O_{12}$(170K)	7.13	1.11	no		2.15	24,000	2,000	12	26
CaF_2(Eu)	3.19	3.72	no	435	1.44	19,000	940	20	4, 8, 13, 27
$CaWO_4$	6.1	1.50	no	430	1.92	6,000	6,000	1	11
$CdWO_4$	7.90	1.21	no	470	2.30	15,000	15,000	1	13, 19, 28
CeF_3	6.16	1.77	no	340	1.62	4,200[e]	27[e]	155[e]	29-35
				300		200[e]	3[e]	65[e]	
CsF	4.11	2.69	very	390	1.48	2,500	2.9[d]	860	5, 13, 36-38
CsI(Na)	4.51	2.43	yes	420	1.84	39,000	630	62	13
CsI(Tl)	4.51	2.43	no	540	1.80	59,000	800[f]	f	13, 28,
						5,400	6,000	1	39, 40
CsI(pure)	4.51	2.43	no	315	1.80	2,300	16	140	13, 41
Gd_2SiO_5(Ce)	6.71	1.50	no	440	1.85	10,000	60	170	13, 42-44
LiI(Eu)	4.08	2.73	very	470	1.96	11,000	1,400	8	13, 45
NaI(Tl)	3.67	3.05	yes	415	1.85	38,000	230	165	13, 28
NaI(77K)[g]	3.67	3.05	yes	303	1.85	76,000	60	1,300	6, 46
NE102A	1.03	10.5	no	425	1.58	10,000	2.4	5,000	11
Pilot U	1.03	10.5	no	425	1.58	10,000	1.36	7,300	11
$ZnWO_4$	7.87	1.19	no	480	2.2	10,000	5,000	2	28

[a] Heavy scintillators emphasized- others included for reference. Data for room temperature unless otherwise noted

[b] Attenuation length (in cm) for 511 keV photons

[c] $C_{14}H_{10}$ (three fused aromatic rings)

[d] cross-luminescence

[e] subject to sample-to-sample variations

[f] 40 ns rise time

[g] Pure (undoped)

Table 2 Heavy Scintillators in Limited Availability

	density (gm /cm^3)	μ^{-1} a	hygro- scopic	λ_{max} (nm)	index refr.	photons /MeV	decay time (ns)	photons /MeV /ns	refs
$BaLiF_3$			no	240		1,800	<1.0[b]	>1,800	20
$Bi_4Si_3O_{12}$	7.13	1.06	no	480	2.06	1200	100	12	12
CdF_2	6.64	1.76	yes	540	1.55	200	10	20	47
CdS(Te)	4.82	2.39	no	640		190	18	11	48
						3,170	270	12	
						13,640	3,000	5	
						total = 17,000		total = 28	
CsBr(80K)	4.44	2.58	yes	250		1,800	1.34[b]	1,340	38
CsCl	3.99	2.79	yes	245, 270	1.64	900	0.88[b]	1,000	38
$KLuF_4$			no	190		170	1.3[b]	130	20
LaF_3 (Ce)	5.94	1.85	no	290	1.7	220	3.0	73	33, 49
				340		1,890	26.5	71	
						90	185	0.5	
						total = 2,200		total = 145	
LaF_3(Nd)	5.94	1.85	no	173	1.7	1,800	6	300	50
Lu_2SiO_5(Ce)	7.4	1.22	no	420	1.82	30,000	40	750	51, 52
$PbCO_3$	6.6	1.16	no	475	1.80, 2.04	180	2.0	90	53-55
						550	15	37	
						70	92	1	
						total = 800		total = 128	
$PbMoO_4$(100K)	6.92	1.22	no			6,000	11,000	0.5	56
$PbSO_4$	6.4	1.28	no	340	1.85	5,500	5, 26, 135	100	57-59
$PbSO_4$ (170K)	6.4	1.28	no	340	1.85	27,000	300	90	59
						41,000	1,500	27	
						total = 68,000		total = 117	
$YAlO_3$(Ce)	5.35	2.24	no	390	1.94	19,700	31	635	60, 61
$Y_3Al_5O_{12}$(Ce)[c]	4.55	2.63	no	590	1.82	11,000	50, 290		62
Y_2SiO_5(Ce)	2.70	4.43	no	420	1.8	45,000	70	640	63, 64

[a] Attenuation length (in cm) for 511 keV photons

[b] cross-luminescent

[c] also known as YAG

Table 3 X-ray excited fluorescence of heavy compounds not available as crystals

	density (gm /cm^3)	μ^{-1} a	hygro-scopic	λ_{max} (nm)	index refr.	photons /MeV	decay time (ns)	photons /MeV /ns	refs
$BaCl_2$	3.90	2.89	yes	300	1.74	8,600	1.2	7,200	66
				300		5,800	58	100	
						total = 14,400		total = 7,300	
$CeCl_3$	3.90	2.85	yes	360		1,800	4.4	410	66
				360		19,600	23	850	
				360		2,100	70	30	
						4,500		>10 μs	
						total = 28,000		total = 1,290	
CuI	5.62	2.04	yes	430	2.35	600	<0.5	>1,200	66
$LuPO_4$(Ce)	6.53	1.43	no	350		200	5	40	66
				350		4,200	23	180	
						total = 4,400		total = 220	
$PbWO_4$[c]	8.2	0.96	no	460	2.3	140	1.7	82	65, 66
						170	10	17	
						110	38	3	
						70		>10μs	
						total = 490		total = 102	
Yb_2O_3	9.17	0.97	no	350		100	<0.5	>200	65, 66
ZnO(Ga)	5.61	2.16	no	385	2.02	15,000	0.7	21,000	11, 68-70
ZnS(Ag)	4.09	2.94	no	450	2.36	49,000	200	250	10, 11

[a] Attenuation length (in cm) for 511 keV photons

[b] Approximate value- subject to uncertainties in the optical depth of the powders used

[c] Values from measurements of powdered samples[66]. Scintillation measurements of crystal samples in excellent agreement[71]. Recently, large crystals have been grown[72].

5. A Method for Finding New Heavy Scintillators Using the Pulsed X-ray Method

We propose a comprehensive, efficient program for finding new scintillators, which includes the following steps:

1 Identification of promising pure and doped compounds using theoretical or empirical knowledge. Since there are many thousands of dense, heavy atom compounds and each of these could be doped with over 50 elements in a variety of concentrations, some guidance is necessary.

2 Preparation of the compounds identified in step 1 in powdered form. Note that in most cases, the chemical synthesis of a powdered sample is considerably easier than growing a clear crystal ≥ 5 mm in size.

3 Exposing the powdered samples to pulsed x-rays and detecting and measuring interesting (bright, fast, etc.) fluorescent emissions. X-rays provide a broad spectrum of energy transfers and produce thousands of excited neutral and ionized molecules. Photons from a UV lamp are not energetic enough to excite all important scintillator transitions (such as the fast crossover transitions in CsF and BaF_2). A synchrotron VUV beam can provide monoenergetic photons over a wide range of energies, but only crystal samples can be used because an ultra-high vacuum is required.

4 Growing small (≥1 cm^3), clear crystals of the few (<1%) of the compounds found to be interesting in step 3

5 Exposing the crystals from step 4 to gamma rays to detect a photopeak and measure the luminosity, energy resolution and timing resolution. The use of a single crystal sample is necessary to include the effects of self-absorption, which can occur in molecules with non-filled electronic shells. Because this self-absorption can seriously reduce the light output, it is said that “the birth certificate of a new scintillator is its gamma-ray photopeak.”

6. Conclusions

1 Theoretical or empirical guidance is vital for a comprehensive and efficient search of new heavy scintillators.

2 There is much work to be done by crystal growers in producing scintillation-quality crystals of compounds with interesting x-ray excited fluorescence emissions.

3 Compared with the tasks of developing a predictive theory and growing large, clear crystals, characterization of powdered and crystal samples is easy.

4 The birth certificate of a new scintillator is its gamma ray photopeak (or monoenergetic charged-particle peak).

5 We may never find the scintillator that is “ideal” for all applications, but the discovery of new scintillators will provide a wider selection of properties for a variety of applications.

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